Synopsis

Al-alloys containing Mg infiltrate spontaneously into porous ceramic preforms in a nitrogenous atmosphere above 700°C The governing process parameters are temperature, alloy composition, atmosphere and preform characteristics (particle size and porosity) In the present work, the role of the process parameters on pressureless infiltration of Al-Mg based alloys into Al_2O_3 preforms have been studied The study has been divided into two sections (1) open conditions wherein the infiltration front is in continuous contact with the furnace atmosphere and (2) sealed conditions wherein the infiltration front is no longer in contact with the furnace atmosphere once the metal billet melts and seals off the front from the furnace ambient Among all the process parameters, Mg in the system, either as an alloy addition or as a powder at the billet/preform interface, and N_2 in the atmosphere have been found to be the most crucial parameters in effecting the phenomenon of pressureless infiltration

The role of Mg has been found to be two fold (1) in initiating the infiltration and (2) in continuing the infiltration In both the stages, the role of Mg is to make the infiltration front free of a passivating alumina layer, which forms due to the reaction of the melt with the residual oxygen in the nitrogen Mg initiates the infiltration by reducing the impervious alumina layer on the alloy billet thereby enabling the alloy to come in contact with the preform Al can also be infiltrated into the Al_2O_3 by keeping Mg at the alloy/preform interface without having to pre-alloy with Mg This method offers flexibility in choosing the alloy composition It has been found that the heating rate has a significant effect on the initiation of infiltration Slower heating rates (<10°C/min) or faster rates of heating (>10°C/min) with an intermediate hold at a lower temperatures (<700°C) facilitate early initiation However, with faster heating rates without any intermediate soaking, the infiltration process is characterised by an incubation period This incubation period appears to depend on the amount of time the sample spends at lower temperatures before it reaches the set temperature and the residual oxygen content of the N_2 The difference in the initiation behaviour as a function of heating rate has been attributed to the difference in the structure and morphology of the surface oxide layers

During the infiltration, Mg evaporates from the infiltration front and getters the residual oxygen thereby making the front free of any passivating oxide layer It has been observed that the infiltration terminates abruptly at a certain height, which is a function of temperature and the Mg content of the alloy The termination phenomenon which has been observed in all the alloys studied, *i e*, Al-Mg based binary, ternary and quaternary alloys is due to the local depletion of Mg at the front This depletion results because the rate of evaporation of Mg at the front is higher than the rate of supply of Mg from the alloy reservoir A way of overcoming the depletion of Mg at the front has been devised, which consists of using a preform whose cross-sectional area is smaller than that of the billet By this technique, a continuous supply of Mg required for gettering the residual oxygen can be ensured through the evaporation of Mg from the annular region between the crucible and the preform However, this technique cannot be extended to the high temperature region (>1000°C) where the matrix formed is itself a composite of AlN/Al Gettering of oxygen can be achieved by the Mg present in the alloy (internal getter) or an alloy containing Mg kept in the path of N_2 gas (external getter) The latter method brings down the residual oxygen concentration in the nitrogen before it reaches the infiltration front thereby making the infiltration continue for longer periods of time even if the Mg content of the infiltrating alloy is very low Also, the use of an external getter lowers the temperature at which significant amount of AlN forms

Based on the understanding of the role of Mg in the continuation of infiltration, it has been possible to devise a technique to getter the residual oxygen completely in order to eliminate the phenomenon of termination In this technique, the infiltration front does not come in contact with the furnace atmosphere after the alloy billet melts and seals off the preform from the furnace ambient This method has been extended to processing in air atmosphere by keeping a small amount of Mg at the billet/preform interface and introducing the sample into the furnace at the set temperature, which obviates the need for a N_2 atmosphere or for Mg in the alloy By using this technique, it has been demonstrated, for the first time, that commercial purity Al can be infiltrated into Al_2O_3 in air at 800°C without applied pressure, with as little as 0 4 wt% Mg enrichment in the metal at the end of the infiltration A number of Al-alloys, *viz*, Al-Mg, Al-Si, Al-Zn, and combinations thereof have been infiltrated successfully into Al_2O_3 preforms in air However, very little infiltration occurs in oxygen while no infiltration occurs in argon In argon, the preforms are discoloured due to the coating

and reaction of Mg with the preform particles The extent of discoloration is a function of the amount of Mg in the alloy or at the interface, and the temperature Thermodynamic and kinetic analyses of the chemical reactions at the front in the Al-Mg-N-O system indicate that AlN is the phase most likely to form on the surface of the melt in N_2 under open conditions Under sealed conditions in air as well as N_2, MgO is likely to form on the melt surface In the case of argon, however, the melt surface appears to be free of any reaction products The role of atmosphere in both the open and sealed conditions is not clear However, it is evident that a critical amount of N_2 is needed for bringing about the phenomenon of pressureless infiltration, though whether its role is limited to the initiation or both initiation and continuation is not obvious

Under sealed conditions, the absence of termination can enable the measurement of the equilibrium height of capillary rise of Al-alloys An attempt has been made to measure the equilibrium height of infiltration with a view to obtaining the contact angle, θ, and surface tension, γ_{LV} The equilibrium height of the alloy containing the lowest amount of Mg used in this study, *i e* , 2%, has been found to be more than 10 mm which is the maximum size of the preform allowed by the experimental set up used It was therefore not possible to estimate γ_{LV} and θ It has been shown, contrary to earlier reports, that the contact angle between Al and Al_2O_3 is less than 90° at 800°C, as indicated by the fact that Al infiltrates into Al_2O_3 under these conditions

Modelling studies have been carried out in order to obtain quantitative information such as the infiltration rate as a function of particle size and the temperature Al-5%Mg, Al_2O_3 preforms of 40% porosity and N_2 atmosphere have been chosen as a typical system The infiltration studies have been carried out in the temperature range of 800-875°C with particle sizes in the range of 0 5-90 μm The existing models based on Poiseuille's flow for capillary rise of non-reactive liquids such as water and organic solvents have been used to predict the infiltration rate as a function of particle size and the temperature The models overestimate the infiltration rate by four to five orders of magnitude Such large discrepancies between the observed and the calculated values are believed to be caused by the presence of AlN on the melt surface and its effect on the rate of infiltration Thus, these models are inadequate to quantify the

pressureless infiltration of reactive metals such as Al It has been observed that the rate of infiltration first increases with decreasing particle size down to 25 μm, and then decreases with further decrease in the particle size to sub-micrometer range The models cannot predict the experimentally observed maximum in the infiltration rate with particle size since the effect of reaction products formed at the infiltration front on the rate of infiltration is not taken into account It is observed that the infiltration rate increases by about a factor of five when the temperature is raised from 800-875°C This increase in infiltration rate is attributed to the decrease in contact angle with temperature